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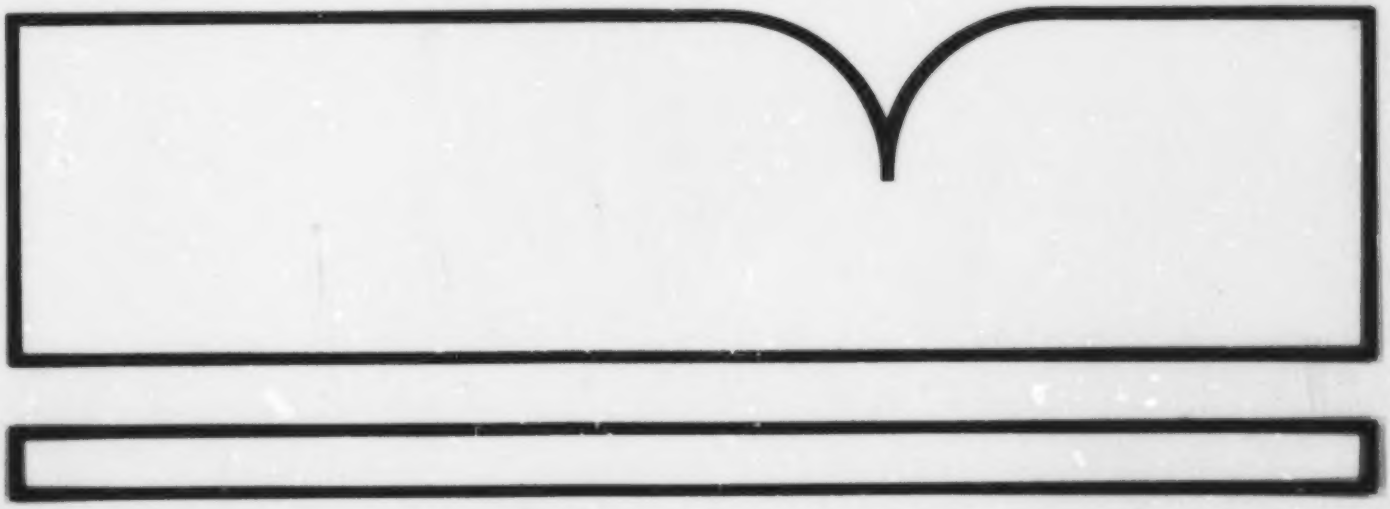

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APPLICATION OF A SEDIMENT DYNAMICS MODEL FOR ESTIMATION OF VERTICAL BURIAL RATES OF PCBs IN SOUTHERN LAKE MICHIGAN

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15. SUPPLEMENTARY NOTES

16. ABSTRACT

The recovery of Lake Michigan from PCB contamination depends on the rates of reduction in PCB input from external sources and removal of PCBs present in the lake system. The loading of PCBs to the Great Lakes should be decreasing. Consequently, if the PCB burden of the lake is also declining, recovery should be occurring.

Because the removal of PCBs from the lake water to the bottom sediments is relatively rapid, sediments could play a role as an ultimate sink for PCBs if the normal accretion/burial process is faster than the rate at which PCBs can diffuse through sediments.

In this paper vertical burial model is developed to estimate the time scale of the burial process. The model is implemented for PCBs in southern Lake Michigan. Vertical sediment dynamics are two-dimensionally interpolated from available literature data. PCB distributions over the southern basin are presented. The burial process is investigated by examining model projections of the hypothetical case where sedimentation continues as normal and there is no new input of PCBs into the lake system.

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Importance of PCBs in Lake Michigan Sediments

PCB residues are observed in the sediments of Lake Michigan. The recovery of Lake Michigan from PCB contamination, as reflected by PCB levels in fishes, depends on the rates of reduction in PCB input from external sources and removal of PCBs present in the lake system. The loading of PCBs to the Great Lakes should be decreasing. Consequently, if the PCB burden of the lake is also declining, recovery should be occurring.

The removal of PCBs from the lake water to the bottom sediments is relatively rapid (residence time $\sim$ 2 years) (1), but PCBs in surficial sediments are resistant to degradation (2) and are recycled into the pelagic food chain by bottom-feeding organisms and resuspension. However, the PCBs in the surficial sediments are gradually buried through the processes of sedimentation.

It seems possible that sediments could play a role as an ultimate sink for PCBs if the normal accretion/burial process is faster than the rate at which PCBs can diffuse through sediments. The mobility of PCBs in fine grained sediment is apparently very low ($\sim 5 \times 10^{-10}$ cm^2 sec^{-1}) and is not expected to compete with the linear net sedimentation rates in the depositional zones found in any of the Great Lakes with the possible exception of Lake Superior (3).

A vertical burial model is developed here to estimate the time scale of the burial process. The model is implemented for PCBs in southern Lake Michigan. Vertical sediment dynamics are two-dimensionally interpolated from available literature data. PCB distributions over the southern basin are presented. The burial process is investigated by examining model projections of the hypothetical case where sedimentation continues as normal but with a zero PCB level.

Sediment Dynamics

Vertical dynamics of the surficial Lake Michigan sediments has been intensively investigated by others using radionuclide profile methods (4,5)

and is described briefly here. A single layer mixed zone model was calibrated with the naturally occurring Pb-210 sediment profile. For calibration, time-invariant sedimentation rates, bulk densities, and mixed zone depths were fitted to the observed profile assuming a constant Pb-210 input rate. The one layer model was found to adequately describe the observed core profiles. The model was then used to predict the shape of the Cs-137 profile given the variable levels of Cs observed in fallout. Predicted profiles matched observed zones exceedingly well. These results appear to be an adequate verification of the utility of the one layer mixed zone model for dealing with the dynamics of burial through the sediment surface.

Vertical Burial Model

The model used here to project vertical burial is essentially the same as used by Robbins and Edgington (4), but extended into the future. PCB is treated as a contaminant strongly bound to sediment, similar to lead and cesium. For the purposes of future projection of vertical burial, the original model can be operationally simplified while retaining the ability to use the original parameterization.

It is assumed that the surface sediment layer, of thickness S, is completely and continuously mixed. Sediment is added to this mixed zone at the surface and mixed zone depth is maintained by loss of material at the base. The mixing rate of material between the mixed zone and underlying sediment is assumed to be zero. For our purposes, there is no need to track the contamination level of material exiting the mixed zone since we are concerned solely with the uppermost sediment layer. Under these assumptions, only PCB in the mixed zone can move upwards. Similarly, we do not need to account for sediment compression. In the simulations presented here mixed zone material is assumed to have uniform and constant porosity (0.9), and solids density (2.45 g/cm^3). Originally, the vertical burial model was implemented using a finite difference approach similar to that used by Robbins and Edgington (4). For the simulations presented here, the model was reprogrammed for use on a PDP 11/70 with analytical solution. The analytical solution is presented below.

For the case where sedimenting material is contaminant-free, the decrease in contaminant surface concentration is directly related to sediment

replacement in the mixed zone, and is first order in concentration, C:

$$\frac{dC}{dt} = -kC \tag{1}$$

Separating variables, integrating over time, and solving for the integration constant given that $C_{t=\infty} = 0$ and $C_{t=0} = C_o$ yields:

$$C_t = C_o \exp(-kt) + 0 \tag{2}$$

The time constant k is the replacement rate of mixed zone material:

$$k = \frac{R}{S} \tag{3}$$

where R = linear sedimentation rate, cm/yr, and S = mixed zone depth, cm. Linear sedimentation rate R can be calculated directly from mass sedimentation rate:

$$R = \frac{\omega}{(1-\phi)\ \rho s} \tag{4}$$

where ω = mass sedimentation rate, $g/cm^2/yr$; ϕ = porosity, cm^3/cm^3; and ρs = solids density, g/cm^3.

For the case where sedimenting material is contaminated at a level C', the differential equation is:

$$\frac{dC}{dt} = -k(C-C') \tag{5}$$

Given that $C_{t=}$ = C' and $C_{t=0}$ = 0, this yields the following result by reduction:

$$C = (C_0 - C') \exp(-kt) + C' \tag{6}$$

where k is as per equation 3. Equation 6 is easily implemented given C_0, C', R, S, and a time increment. The concentration units of C' must match those of C. Data in the usual terms of µg contaminant per gram dry weight of sedimentary particulate (C_{ss}) must be corrected for mixed zone bulk density, viz:

$$C' = C_{ss}\ (\phi-1)\rho s \tag{7}$$

Physical parameters used to implement the burial model were obtained from John Robbins (6). It should be mentioned that these values represent data available in 1978; some of the values may have been revised after subsequent cores were analyzed.

The fact that the burial model is an extension of the model used to derive the parameters ω and S speaks strongly for the validity of their use. It is certain that the single layer model is simpler than reality. In nature, the mixing rate of sediments is a variable function of depth and net

sedimentation is the difference between gross sedimentation and resuspension. But since the parameters ω and S are fitted to observed data (Pb-210) and demonstrably predict surface sediment dynamics (Cs-137), there is good reason to expect continued validity in predicting future dynamics. Underlying this is the further assumption that ω and S can be considered time-invariant; this appears to be true and is argued at length elsewhere (7).

Current PCB Concentrations in Surficial Sediment

PCB analysis was conducted on sediments from 30 sites obtained during three cruises in southern Lake Michigan in the summers of 1978 and 1979. In most cases, cores were obtained with a 9 x 25 cm box corer. Sediments were obtained with a ponar sampler at a few locations. The cores and ponar samples were sectioned in the field and stored in glass jars at 4°C until analysis was conducted. The data presented here represents the upper section (0.5 or 1 cm) of the sample. The sediments were extracted by continuous steam distillation/extraction followed by cleanup by elution of the extract in hexane through a column containing anhydrous Na_2SO_4 (2 g) over 10% deactivated alumina (5 g) over acid-washed Cu filings (5 g) (ref. 8, for details). The extracts were analyzed by electron capture gas chromatography using a Hewlett-Packard Model 5830A or 5840A instrument equipped with 60 m SCOT column (SP 2100) or 50 m WCOT column. PCB concentrations were calculated by comparison to Monsanto Aroclor mixtures or EPA Aroclor standards using multiple linear regression analysis (9).

Implementation and Results

For the purposes of this work, a 33 x 39 grid was defined over the southern basin of Lake Michigan. The grid is oriented E-W along latitude 41°30'N with the origin being 41°30'N by 88°0'W, and each grid cell side equal to 5 km. (This scale corresponds to centimeters on NOAA Map 7 which is a 1:500000 map.) 807 of the grid cells represented lake surface, with the remainder representing land surfaces. Each of the over-water cells was parameterized for the burial model and run independently.

As noted above, the physical and chemical parameters were obtained from different core sets. Neither set covers the whole southern basin. The U. of Michigan (physical data) stations are biased towards the eastern shore

and the U. of Wisconsin stations (PCB data) are concentrated near Waukegan and the basin center. Figure 1 shows the station locations, individual, and combined geographic coverages of the data sets.

Each of the 807 relevant grid cells was assigned a value for sedimentation rate, mixed zone thickness and current PCB concentration (Table 2). A modified inverse-distance-weighted averaging alogrithm was used to perform the two-dimensional interpolation (11,12). Figure 2 shows the distribution of ω and S in contour plot display. The grid cell values were averaged and integrated by summing. Mean sedimentation rate for the area of interest was 26.7 $mg/cm^2/yr$ which represents 5.4×10^6 metric tons per year over the area of interest. Average mixed zone thickness was found to be 1.38 cm, corresponding to a total mixed zone volume of .278 km^3. The PCB distribution was generated in a similar manner (Figure 3). PCB concentration over this area averages 61 parts per billion (wet weight). The mixed zone volume and PCB concentration data were combined and integrated cell-by-cell to yield a total mass in the mixed zone of 2760 kilograms.

It must be emphasized that these averages and integrations are based on sediment samples which are not evenly distributed over the lakes. The values above are extrapolated to the whole southern basin but in fact only reflect the area which was sampled. Comparison with Cahill (13) shows that these stations are biased towards depositional and transitional regions, leading to inflated estimates of basin-wide sedimentation rate. This consideration does not affect the following analysis, however, because cell-by-cell simulation will be valid for the sampled areas.

The burial rate of each cell is determined by R and S, as shown above. A simple parameter which characterizes this is the 50% response time:

$$t_{1/2} = - \frac{\ln (.5)\ S}{R} \qquad (8)$$

The values for $t_{1/2}$ were computed for each cell and are displayed in Figure 4. A histogram of the $t_{1/2}$ distribution is also included showing two modes at about 5 and 10 years, respectively. The mean $t_{1/2}$ is 7.5 years.

Simulation was set up for the best case clean-up situation. In these simulations, the PCB concentration in sedimenting particulates is set

to zero. Output grids were generated and displayed for 0, 5, 10, and 20 years (Figure 5).

Comparison of the observed sediment PCB concentrations with the predicted sediment response time can provide information on PCB sources and trends in PCB concentration of the depositing sediment. If diffuse sources are of primary importance, or if suspended sediment is well mixed before permanent sedimentation, a correlation may exist between these two distributions. Four scenarios are presented: I) Deposition of sediment of constant PCB concentration has occurred sufficiently long for the PCB concentration in the surface layer to reach a maximum level. The concentration in the surface layer would be constant throughout the basin and equal to the concentration in the accumulating sediment. II) The deposition of sediment of constant PCB concentration is ocurring, but the concentration in the surface layer has not reached a maximum level throughout the basin due to dilution by mixing with underlying sediment. In this case, an inverse relationship between PCB concentration and $t_{1/2}$ is expected (low PCB concentration where $t_{1/2}$ is long). III) The PCB concentration has reached a maximum level as in (I), and the concentration in the surface sediment is now decreasing due to a decrease in the level of PCB contamination of the depositing sediment. In this case, a direct correlation between surface layer PCB concentration and $t_{1/2}$ is expected because concentrations would decrease most slowly in areas with the longest response time. Also, a sub-surface maximum is expected. IV) The level of PCB contamination of the depositing sediment began to decline before the concentration in the surface layer had reached a maximum throughout the basin. In this case, areas where $t_{1/2}$ is long would not have reached a maximum, and the decrease in concentration would also be slow. A sub-surface maximum might occur. However, areas where $t_{1/2}$ is short would have reached higher surface concentrations in the past, and now the concentrations would be decreasing more rapidly. A sub-surface maximum would be expected. Consequently, the relationship between surface layer PCB concentration and $t_{1/2}$ would be complex for scenario IV.

The two distributions of interest here are shown in Figures 3 and 4. The observed [PCB] distribution and the computed 50% response time for the sediment mixed zone appear to be positively correlated over almost all of the sampled area. As described above (scenario III), this implies that the

surface sediment PCB cocentration has reached a maximum and is now decreasing over most of the basin. This conclusion is strengthened by the fact that [PCB] is well correlated with mixed zone depth (+.57).

Higher PCB levels also tend to be found in depositional than in non-depositional zones, suggesting that much of the PCB gradient is due to lateral transport of contaminated sediment. A striking exception is the very high [PCB] region near the south-southeast shore (>100 ppb). This region is not associated with either a high or a low response time prediction. A characteristic of this area is a relatively low mixed zone depth and a thin layer of fine-grained sediment overlying sand and glacio-lauclustrine clays (low net sedimentation). The possibility of a point source is indicated here. It is not possible to differentiate between a local point source (such as Gary or Michigan City) or a remote point source because lateral sediment transport is not accounted for by this model.

Simulation of Future PCB Burial

The projected response of Lake Michigan sediments to burying surficial PCBs is shown in Figure 5. An obvious point to be made is that "hot spots" are predicted. The actual development of these areas may not occur as shown due to lateral transport. It does seem certain, however, that the sediment PCB distribution is and will remain quite heterogeneous. Note that and S are correlated. The areas with slowest response are those with moderate sedimentation and mixed zone thickness. Extreme mixed zone depth appears to be largely offset by high sedimentation, and doesn't lead to extreme retention. The reverse is probably not true: areas with low sedimentation and mixed zone depth are mostly "scoured" and will probably have a response time at least as fast as shown.

Projected average PCB concentrations over the basin are shown in Figure 6. Although different areas have markedly different response times, the lake can be roughly thought of as a single reactor with a 50% response time on the order of 5-10 years. The single mixed reactor model breaks down after approximately four half lives (30 years), but the explicit model probably loses validity by then due to lateral transport.

Conclusions

1. A heterogenous distribution of PCBs was found in the southern basin of Lake Michigan. This was expected and is anticipated to remain heterogenous. Great care is advised in sampling sediments to establish [PCB] time trends, since large variations can be expected within a few kilometers.
2. An average 50% response time of about 5-10 years is expected for the sediments of the whole basin.
3. It appears that the surficial sediments over most of the southern basin have reached their maximum PCB concentration and are now decreasing in concentration.
4. One or more significant point sources are indicated in the south reach of the lake. Further measurements could be of great value, although there is a great deal of difficulty obtaining good samples from this region.

Acknowledgments

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TABLE 1. PCB concentrations in surficial Lake Michigan sediments. Concentrations reported on a dry weight basis.

Station Location* km East	North	[PCB] ng/g
19.0	77.5	3.3***
27.0	83.5	9.3
27.0	110.5	3.1
28.0	91.0	14.2***
32.5	74.0	67.0***
34.0	83.0	13.7
39.0	119.0	13.8
41.0	101.0	68.9
47.0	92.0	13.2
47.5	72.5	2.1
47.5	110.5	18.9
49.5	64.0	1.0***
58.5	102.0	74.5**
58.5	146.0	23.0
61.0	101.0	43.9***
61.5	64.0	25.8
62.5	83.5	48.8
70.5	65.0	6.1
73.5	71.5	16.9
75.0	92.0	69.4
76.0	167.0	52.3
81.5	136.0	91.0
85.5	131.0	69.7*
88.5	37.0	201.1
93.0	90.0	104.0
93.0	108.5	103.3
101.5	55.0	103.1
101.5	195.0	17.5*
102.5	75.0	120.0
119.0	135.0	105.0
128.0	170.0	7.1

* Locations in km east and north of 41°30'N by 88°0'W.

** 1-2 cm sample

***Ponar sample

TABLE 2. Physical data used to implement sediment dynamics model (from Robbins, 6). Note that this data is preliminary.

Station Location* km East	North	Mass Sedimentation Rate ($mg/cm^2/yr$)	Mixed zone thickness (cm)
106.0	203.8	59.0	NA
114.3	203.8	69.0	NA
120.0	203.8	52.0	NA
112.8	184.8	75.0	0.5
108.5	185.3	35.0	1.0
104.5	166.5	75.0	0.5
119.3	166.5	83.0	2.0
127.8	166.5	20.5	1.0
60.5	72.0	15.0	1.6
80.5	147.0	NA	0.3
93.7	147.0	57.0	1.0
122.0	148.3	14.0	2.0
135.3	148.3	95.0	3.5
81.0	128.8	13.0	2.0
95.0	128.8	73.0	2.0
108.5	129.8	74.0	1.0
122.5	129.8	76.0	2.0
136.0	130.5	102.0	1.5
95.5	108.8	9.0	3.0
109.3	109.3	11.0	3.0
113.3	115.0	30.0	2.0
102.8	120.0	25.0	1.4
123.3	110.0	31.0	3.0
128.8	107.8	33.0	2.0
136.0	110.5	26.0	3.0
38.3	91.0	23.0	0.5
55.5	91.0	35.0	1.0
82.8	92.3	9.0	1.0
96.0	92.8	17.0	2.0
110.0	93.3	53.0	2.5
112.0	86.0	64.0	3.7
126.0	80.0	46.0	0.5
98.8	73.8	20.0	2.0
108.3	73.8	35.0	2.0
90.5	53.8	18.0	1.0
100.0	54.5	29.0	1.0
97.8	43.3	22.0	2.5
123.3	93.3	74.0	2.5

* Locations in km east and north of 41°30'N by 88°0'W.

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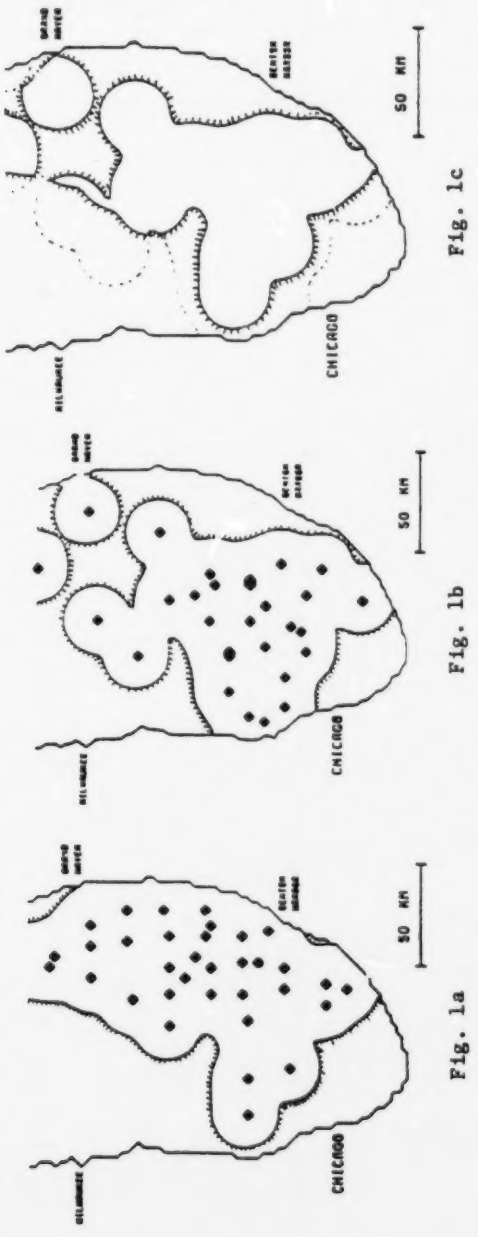

Fig. 1c

Fig. 1b

Fig. 1a

Figure 1. Data set coverage over the southern basin of Lake Michigan. 1a) Sediment dynamics model coverage (6). 1b) Surficial (PCB) sample coverage. 1c) Combined coverage.

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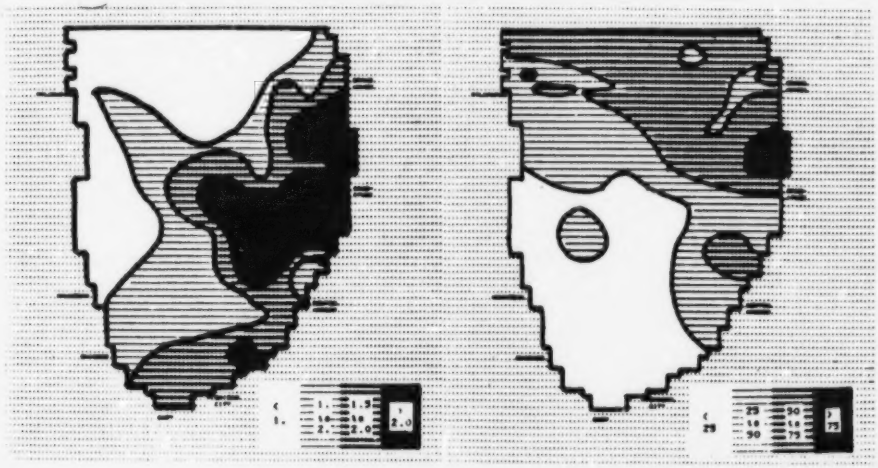

Fig. 2a. Mixed zone depth, cm.

Fig. 2b. Sedimentation rate $mg/cm^2/yr$.

Figure 2. Sediment dynamics model parameters.

Figure 3. PCB concentration in surficial sediment, ppb.

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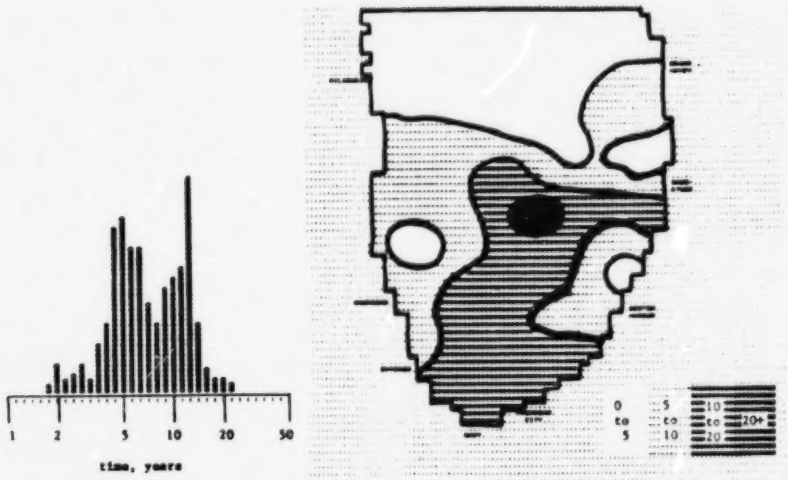

Figure 4. 50% response time for sediment in the surficial mixed zone, years.

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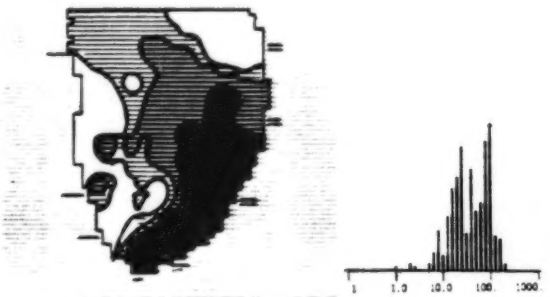

Fig. 5a. Current surficial (PCB) concentration, ppb. Contour intervals are identical to Figure 3.

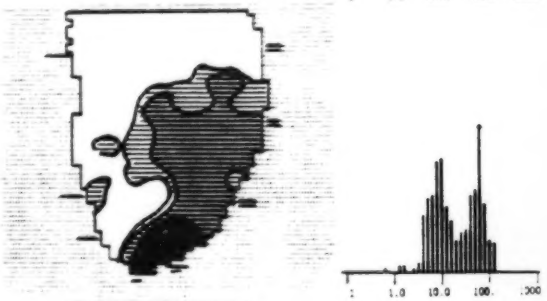

Figure 5b. Best case projection; replacement of surficial sediment with uncontaminated sediment for 5 years.

Figure 5c. 10 years.

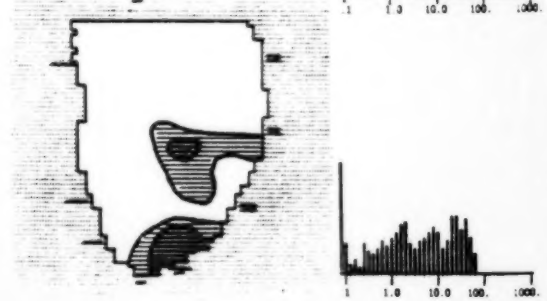

Figure 5d. 20 years.

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TOP OF IMAGE AREA
DROPPED HEAD: BEGIN SECTIONS HERE

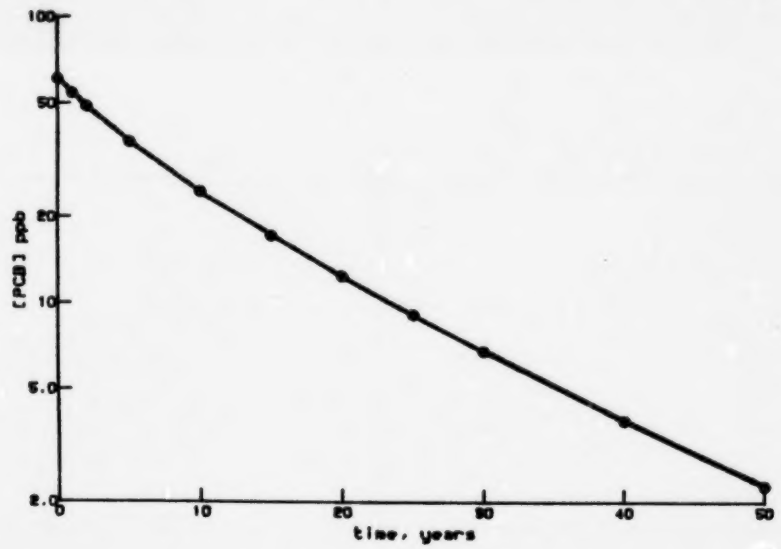

Figure 6. Best case projection mean surficial (PCB). This represents the fastest decrease which can be expected due to vertical burial.

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